GEOCHRONOLOGY, AND ISOTOPIC DATA BEARING ON DEVELOPMENT OF THE CONTINENTAL CRUST*

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Prologue

I will present some ideas and problems regarding the evolution of the crust of the earth and will also attempt a prognosis of the application of isotopic techniques to the understanding of geological processes. This dissertation might well be entitled "Pride and Prejudice"; it will be my implicit assumption that the use of physicochemical techniques and principles, when used with a sound geologic knowledge, has and will help solve important existing problems and will also create new problems and new fields which are themselves an integral part of the science of geology.

The close dependence of geology on the other physical sciences was emphasized 159 years ago by Sir James Hall [1812], who, in commenting on the lack of success of his contemporaries in explaining the origin of mountains, said—"One principal cause of this failure seems to have lain in the very imperfect state of chemistry, which has only of late years begun to deserve the name of a science. While chemistry was in its infancy, it was impossible that geology should make progress;

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since several of the most important circumstances to be accounted for by this latter science are admitted on all hands to depend on principles of the former."

Sir James Hall also indicated that he abstained from further pursuit of the problem which had excited him in deference to his eminent colleague, Dr. Hutton, and did not resume his investigations until after Hutton's demise. Such gentlemanly deference is no longer characteristic of science, and it is at this point that we depart from Sir James' good example. At the present time the variety and intensity of work is causing continuous revision of our ideas, and today's speculation is often assaulted by tomorrow's fact.

The application of the methods of classical nuclear physics to geologic problems started soon after the discovery of radioactivity by Henri Becquerel in 1896. The use of long-lived natural radioactivities in determining geologic and cosmic time scales was pursued by a number of distinguished workers including Boltwood, Hahn, Rutherford, Soddy, and Strutt. Many of the most advanced researches pursued today were considered and attempted by these workers during the early 1900's. Variations of lead isotope abundances were first reported by Aston [1933].

A series of three classic papers by A. O. Nier appeared in *The Physical Review* in 1939 and 1941 on the variation of isotopic abundances due to radioactive decay and the measurement of geologic time. With the appearance of these works, the modern field of isotopic geochronology was born. Throughout much of this time, Arthur Holmes maintained and stimulated the geologic application of radiometric ages. There was a lull for about ten years followed by an explosion of activity.

Men like Inghram, Urey, Brown, Thode, Houtermans, and Gerling, Tuzo, Wilson, and Gentner were looking for new frontiers and directed their students and colleagues to exciting problems of cosmic and geologic importance. This choice may have been partly based on an aversion to "big machines," but the techniques which were developed during the period 1941 to 1951 permitted the accurate routine measurements of quantities of the order of 10^{-11} gram. Concurrent with the development of modern geochronology the study of the abundance variations of stable isotopes has also flourished, particularly due to the activities of H. C. Urey and S. Epstein. The application of these methods to geology by a group of half-breeds who were on the borderland of physics, chemistry, and geology has led to the development of a branch, not a technique, of the earth sciences. There are no old men practicing the art, although there are admittedly some who are prematurely grey and balding.

The great activity in the study of nuclear phenomena applied to the study of geologic problems has resulted in some dislocations within the profession and caused some unhappy conservatives to consider this work more or less outside the legitimate field of geology. The practitioners, often referred to as knob twisters or black box operators [J. Hoover Mackin, 1963], are also relegated to a caste of doubtful legitimacy. There have been excessive and erroneous claims made by some isotopic enthusiasts which have caused ill will, but I suppose this is a trait of enthusiasts in general. There appears to be some doubt expressed in some circles over the need for quantification in the science. That is a moot point. The real need is for understanding earth processes; and whatever methods may be applied with advantage will, of necessity, be used. But, in point of fact, the revolution, which is discomfiting to some professionals, is over. The knowledge which is being obtained is in the process of becoming an essential part of the science of geology. These problems range from the evolution of the atmosphere to the evolution of the crust of the earth and the time scale of the solar system and nucleosynthesis.

Methodology

The methods of geochronology are physical and chemical, and the problems and materials are geological. All of the principal dating schemes (see Table 1) are based on the radioactive decay of a

Table 1

PRINCIPAL DECAY SCHEMES USED IN GEOCHRONOLOGY FOR TIMES GREATER THAN 10^5 YEARS

Parent		Daughter		
U^{238}	$\rightarrow$ intermediate daughters $\rightarrow$	Pb^{206}	He^4	1/6.5 Ae
U^{235}	$\rightarrow$ intermediate daughters $\rightarrow$	Pb^{207}	He^4	1/1.03 Ae
Th^{232}	$\rightarrow$ intermediate daughters $\rightarrow$	Pb^{208}	He^4	$\frac{1}{20}$ Ae
K^{40}	$\rightarrow$	Ar^{40}		
K^{40}	$\rightarrow$	Ca^{40}		
Rb^{87}		Sr^{87}		
Re^{187}		Os^{187}		

long-lived parent to produce an accumulated stable daughter product. Assuming that the amounts of parent isotope and daughter isotope may be measured with the necessary accuracy, there is a need for other physical constants. These are the half-lives of the radioactive species. In all of the intercomparisons of the various dating methods on minerals, it has been possible to obtain, often well before purely laboratory methods, rather good determinations of these constants. But in order to obtain what are in principle "absolute dates," it is necessary to have refined half-lives determined by direct methods. This necessity still exists, and some of our understanding of geologic phenomena awaits further refinement in the determination of absolute decay constants. Dates by two decay schemes are not directly comparable with each other. This is true for all of the decay schemes listed in Table 1 [Aldrich and Wetherill, 1958]. The uncertainties for U^{235} and U^{238} while small are very significant because of the systematics in this dating method [Wasserburg, Wetherill, Silver, and Flawn, 1962]. The Rb^{87} decay constant is still far too uncertain for the precision needed [see Leutz, Wenninger, and Ziegler, 1962] and the Re decay constant should hardly be discussed.

A variety of ages can be obtained in studies on mineral systems from a single rock fragment. This dispersion can be found even within one mineral species by a single method and is due to differential effects of diffusion, weathering, and metamorphism. The ages obtained will have an uncertainty in meaning which, thus, far exceeds the analytical error. Even in cases of very consistent data, discrepancies exist between the various methods, and it is not always possible to distinguish between regular continuous (?) diffusion loss and errors in decay constants. For example, if the size distribution function in the fine size region ($\lambda D/a^2 \gg 1$) of mosaic units in crystals in igneous rocks is relatively constant from macroscopic crystal to crystal, then diffusion losses would be very regular and would not be easily discernible from a decay constant error. With these difficulties in mind, is still possible clearly to resolve events by the Sr^{87}-Rb^{87} and Ar^{40}-K^{40} methods which differ by 100 million years at an age of 1000 million years under favorable circumstances [Zartman, 1963]. The resolution ability of a dating method appears to be related to parent-daughter systematics. This exists for the Sr-Rb system but is particularly true for the coupled U^{235}-Pb^{207} and U^{238}-Pb^{206} systems. The theoretical basis for such systematics (in the absence of intermediate daughter loss) has been given by Wetherill [1956], Tilton [1960], and Wasserburg [1963]. The result is that for this coupled system, regardless of the details of the history of daughter or parent loss, the precise age may be found by

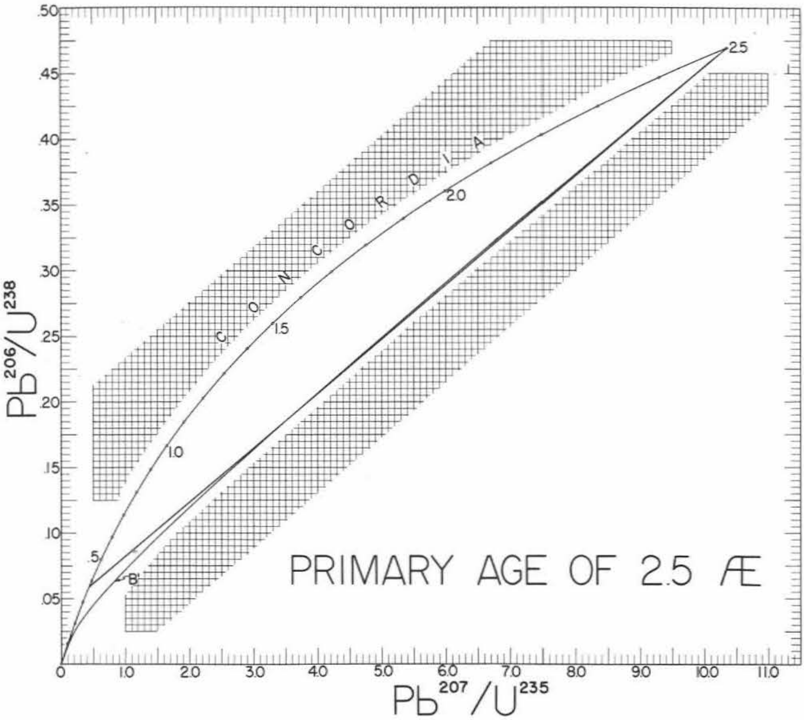

FIG. 1(*a*). Daughter-parent systematics. Lead evolution diagram for the case of continuous lead diffusion loss controlled by radiation damage (B'). The straight line is a tangent to B' at the concordia curve. The linear relationship is always true in the neighborhood of concordia regardless of the law of stable daughter loss.

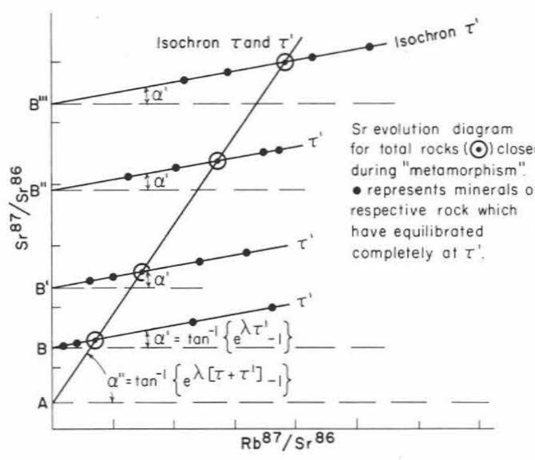

FIG. 1(*b*). Strontium evolution diagram for total rock systems and mineral phases showing the systematics for a two-stage history.

extrapolation of a very linear array of experimental data. This is illustrated in Figure 1*a*. Silver has shown [1963*a*, *b*] in work on different zircon fractions from a given rock that it is possible to obtain remarkably linear arrays and it appears possible to resolve events differing by 60 million years (m.y.) in 1700 million years. This work appears to offer the greatest resolution which can be obtained by isotopic means. Daughter-parent systematics of a less general type are also found for the Rb-Sr system as shown on Figure 1*b*. In general, on both a local scale (hand specimen, outcrop) and on a regional scale, the isotopic ages which are obtained will have a dispersion which limits our ability to define simultaneity. In Precambrian terranes the dispersion of ages due to secondary effects and to intrinsic resolution of the methods used causes us to consider dates in a time band as coherent, when in fact they represent distinct events over a duration of time comparable with a good part of all of the Phanerozoic. This myopic vision may force us to recognize some great geologic truth which is confused in the details of the more recent record or it might also lead us astray. The desire to believe in magic numbers—precise episodicity in magmatism and metamorphism—is at present rather strong in the trade and encourages the label of contemporaneity even when the ages are well resolved. This is further confused by the varying quality of the different studies reported in the literature, so that ages which are a "trifle" low for particular magic numbers are assumed to be due to daughter loss and are automatically upgraded in their assignment. This general problem has been previously discussed in the literature from different viewpoints [Gilluly, 1949, 1963; Wasserburg, 1961; Wasserburg, Wetherill, Silver, and Flawn, 1962]. With the preceding *caveat*, I will proceed with the main topic.

Introduction

In understanding the evolution of continental masses, the question of the growth of continental bodies through geologic time is of fundamental importance. It appears well demonstrated that the continental masses are intrinsically unstable due to erosional processes, and hence accretionary processes must be active in order to maintain regions of positive relief over long periods of time. With regard to the hypothesis of continental evolution, it is necessary to establish whether there exist major subdivisions which have meaning in terms of evolutionary characteristics and to determine whether the geologically younger parts of the continental mass represent the addition of new material or are in fact the product of metamorphism of older, preexisting provinces or materials derived therefrom. In addition, it is of prime importance to identify new material, derived from depth, which is being added to the crust. The continental and oceanic crusts are the two obvious distinctive features. Seismic and volcanologic observations indicate that the sources of lavas on some oceanic islands are from a depth associated with the (seismic) upper mantle [Eaton and Murata, 1960]. Because of the thinness of the oceanic crust and sediments, it is usually inferred that the oceanic volcanic magmatic rocks represent fractionated upper-mantle material relatively uncontaminated by crustal material. The implicit assumption which usually follows from this is that the oceanic mantle is compositionally a uniform layer. This presumed simplicity may be partly a reflection of our lack of observations but is also deeply tied to the question of the age and stability of the ocean basins themselves and the consequences of crustal evolution and of continental drift. The absence of a thickness of deep-sea sediments as estimated from modern sedimentation rates and the age of the earth may indicate some processes of transformation which could produce an upper mantle of considerable chemical complexity. By contrast with what is surmised about the oceanic crust, the continental crust is known to be of great complexity in both structures and rock types, and it has not been possible to attribute directly the magma sources of continental rocks to the mantle. However, we are able to sample materials in the continental crust of great age, but so far no ancient samples of oceanic crust have yet been recognized. The oldest igneous material from oceanic islands is only 650 million years, while the oldest known sediments are only Cretaceous in age. By contrast, exposures of continental crust as old as 2700 million years are quite common. It follows that we can only investigate the products of relatively recent and intense magmatic activity in oceanic regions [Menard, 1964] which are derived from a presumed "simple" region and from continental rocks which represent a wide range in age but which may often represent a complicated, hybrid history.

I will give examples of isotopic studies made in both types of regions and attempt to indicate what additional knowledge they have contributed. My emphasis will be on the Precambrian because it is in our understanding of this time region that these techniques have had their greatest impact.

The Grenville Province

The Grenville province is a large subcontinental segment of the Canadian shield (see Figure 2). This province was defined principally on the basis of structural and lithologic criteria which distinguish it from the adjacent Superior province which bounds it on the west. The Grenville province contains extensive areas of marbles intermingled with a variety of gneisses, highly metamorphosed sedimentary and volcanic rocks, and numerous granites. The structural relationships within the Grenville province are often exceedingly complex, and the granitic rocks are commonly gneissic and concordant bodies in a

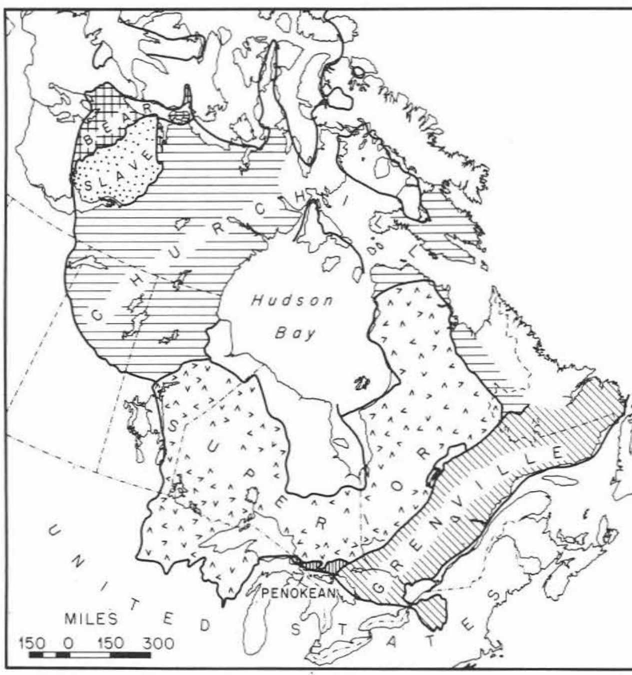

FIG. 2. "Structural" provinces of the Canadian Shield [Stockwell, 1962].

generally migmatitic terrane. The structural trends of the Grenville province transect the structural trends of the so-called Archean and Proterozoic sedimentary rocks of the Superior province which are characteristically of low metamorphic grade. The zone bounding these two provinces is called the Grenville front and is in part a zone of faulting and in part a region of metamorphic transition. The metamorphic and igneous complexes of the Adirondack mountains and the New Jersey highlands are similar in character to that found in the "typical Grenville" province areas and have naturally been associated with this province. The eastern boundary of the Grenville province is uncertain, to say the least, but on lithologic grounds may extend into eastern Canadian Appalachia. The further extent of this great province into the United States has been hazarded by some authors based on the fundamental theorem of tectonics which says that orogenic belts continue along their strike.

Very thorough and sophisticated geologic investigations have been made in many of these areas which have cast great light on the stratigraphy, metamorphism, and local geologic history, but in general no specific ideas have been put forth which would permit a genetic interpretation that could be of use in predicting the character and scale of this province "in the large."

Some measurements of isotopic ages within the Canadian Grenville were obtained which lay within a time band of between 800 million years to 1150 million years. As more measurements were made, it became apparent that the "ages of rocks" within the Grenville province were characteristically in this interval, and that this was distinctively younger than the ages of around 2300 million years which appeared within the Superior province [Leech, Lowden, Stockwell, and Wanless, 1963; Lepp, Goldich, and Kistler, 1963]. Age measurements in the Adirondacks and New Jersey Highlands [Tilton *et al.*, 1960; Long, 1961] also gave ages between 800 to 1150 million years. These data showed that the complex structural and lithologic criteria which were originally used to define the Grenville province also appeared to define a time zone. In Maryland, the Glenarm series and the crystalline rocks which underlie them were of uncertain stratigraphic age. These rocks lie to the east of the major folded Appalachian structures. Age determinations indicated that they were metamorphosed during early Paleozoic time ($\sim$350 million years), but more complete investigation by Tilton, Wetherill, Davis, and Hopson [1958] showed that the zircons of the Baltimore gneiss indicated an age of 1150 million years, suggesting that part of the deformed basement of the Appalachian mountain belt consisted of gneisses which appeared to be time-correlative with the metamorphism within the "classical" Grenville province. This was a major discovery in its own right since it showed the possibility of recognizing a time of primary crystallization through later periods of metamorphism [Wetherill, Kouvo, Tilton, and Gast, 1962].

Other investigations showed rocks of about 1000 million years in exposures in North Carolina and in some basement cores from Ohio [Tilton, Wetherill, Davis, and Bass, 1960]. In central Texas a large exposure of a Precambrian metamorphic and igneous complex was found to be of about 1050 million years [Aldrich, Wetherill, Davis, and Tilton, 1958]. An age study of the exposed Precambrian and of basement cores in an E-W section across the state of Texas showed what appears to be a continuous band of rocks with ages of about 1000 million years which are bounded to the north by older metamorphic and igneous rocks (1200? to 1450). The regions adjacent to this belt are currently being investigated [Wasserburg, Towell, and Steiger, 1965] and show a distinct age jump from 1090 to 1400 million years between El Paso and the southern end of the San Andres mountains—ages of 1400 million years appear consistently in a N-S traverse from White Sands to Albuquerque.

In some localities in the Precambrian of Texas, the lithologic types and structural characteristics are comparable with what are found in the Grenville province, but in many places the rocks can hardly be correlated on such a basis. It is, obviously, plausible to correlate these rocks which were crystallized or metamorphosed at ~ 1000 million years and to make this an extension of the Grenville province. In doing so we obtain a belt of fully continental dimensions (Figure 3). However, this is no longer the Grenville province as defined by lithologic and structural criteria but a continuous (?) region which was subject to contemporaneous (?) crystallization or recrystallization $\sim 10^9$ years ago. It is inferred that this event, or series of events covering about 250 million years, corresponds to an orogenic episode. The term *event* was introduced into the literature to distinguish whatever it is that happened from more specific geologic phenomena. We now frequently refer to regions which show time correlative "dates" as orogenic provinces. This usage, while most probably correct, is an inference. In simple cases the ages which are obtained by the various methods determine a time since the mineral was a closed system with regard to the parent-daughter isotopes. This can, in complicated (but common) circumstances, have very varied meanings as far as "age" is concerned. Assuming that the data are sufficiently coherent so that the age may be taken as representing the last period of metamorphism or intrusion, it is important to make the distinction between these events and orogeny in general. The classical criteria for tectonic activity are structural-stratigraphic, and the time-correlative terms, based on radioactive decay schemes which are now being obtained and used, may have a very different meaning. As emphasized by Gilluly [1963], "radiometric dates yield, not the times of orogenic activity, but of those orogenic episodes which were accompanied by plutonism. Judged by tectonic criteria, these episodes were not more intense than many orogenic episodes not so accompanied." He further states that "there is no obvious correlation of tectonics with metamorphism or with granitic intrusion, though both metamorphism and granite are obviously associated with tectonism." However, I would even prefer to remain uncertain as to the latter correlation. It may be that some of the less tangible "events" which have been reported, such as the

FIG. 3. The belt of igneous and metamorphic rocks which have ages typical of the Grenville time band, which is here taken to be 900 to 1200 million years. Other ages are only shown to indicate the boundary.

so-called 1350 to 1450 million year orogeny, represent tectonic episodes without broad-scale plutonism and (obvious) metamorphism. More recent studies suggest that even the ghostly 1350 to 1450 million year event has extensive associated plutonism.

We have seen from the preceding example that isotopic ages have independently confirmed some of the better-defined structural provinces.* They have also, in some cases, completely upset classifications in Precambrian terranes. The isotopic ages now permit another means of studying orogenic episodes in the Precambrian, but this new technique measures something which is quite different than has been used previously as a basis for describing orogeny. While confirming the coherence of the Grenville province, it has, using a completely different criterion, also added to it a mass of highly varied geologic materials several times its original size. There appear to be even more widespread outliers of this 1000 million year episode: an isolated 1.06 AE rapakivi pluton intrusive into 1.7 AE metamorphic rocks in Nevada, diabase sills in the Sierra Ancha of Arizona and pegmatites in the San Gabriel Mountains of California, 1 AE intrusives at the southern tip of Greenland; and if one really wishes to think big, Grenville ages as far as Norway and Sweden [Polkanov and Gerling, 1961; Kulp and Neumann, 1961]. It is clear that at this rate, the tail may well wag the dog and that what is needed to utilize properly the geologic "age mapping" in the Precambrian, which will undoubtedly be pursued for the next 10 to 20 years, are some theories of orogenesis that can make consequential predictions which are testable by observation and which may prove a format that will permit interpretations to be made; preferably, theories which can correlate the classical structural stratigraphic descriptions with the isotopic ages. This point can be best emphasized by considering the most modern texts on historical geology. One such book recently published, titled *The History of the Earth*, devotes 97 per cent of its contents to a description of 13 per cent of geologic time. This is with good reason; the Precambrian *is* prehistoric in the sense of paleontologic-stratigraphic techniques. And if in fact we were today to expand the coverage to be commensurate with the periods of time involved, while we would have a much thicker book (with many blank pages), the break in methodology would still be seen with the appearance of Olenellus. But the refined morphologic observations which describe parts of earth history (with far finer resolution than absolute age measurements) have not yet provided a genetic-theoretical basis for bridging the gap with "Prehistoric" geology. It is to this point that significant efforts must, in my opinion, be directed.

We will now jump from the broad continental-scale province to

* In the preliminary version of the new tectonic map of Canada (Stockwell, A Tectonic Map of the Canadian Shield, Figure 2, The Tectonics of the Canadian Shield RSC Spec. Pub. No. 4, edited by J. S. Stevenson), it may be seen that the provinces appear to have taken on a strongly time-orogenic character as compared with the previous map. The classification seems to now be based more on a "time" basis than on structural criteria.

consider the boundary between the younger Grenville and the older Superior province within the region delineated on the Canadian shield. This boundary zone is a natural place to determine whether the geologically younger belts represent the addition of new material or are, in fact, the product of metamorphism of the older preexisting geologic provinces. An investigation was made by Grant [1964] across a segment of the Grenville Front south of Lake Timagami, Ontario. By careful field mapping, he was able to demonstrate that the metamorphic transition, which defines the front in this region, is unfaulted

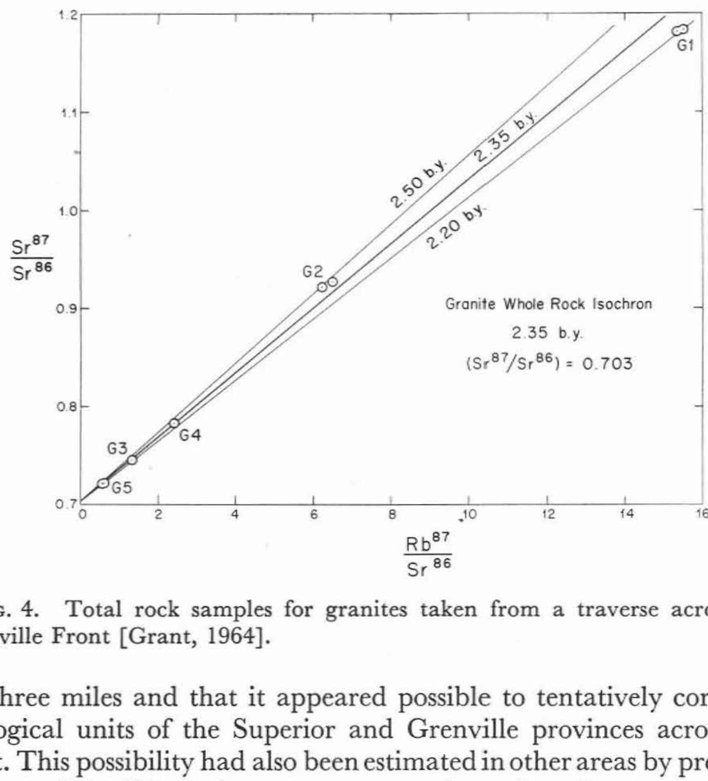

FIG. 4. Total rock samples for granites taken from a traverse across the Grenville Front [Grant, 1964].

for three miles and that it appeared possible to tentatively correlate geological units of the Superior and Grenville provinces across the front. This possibility had also been estimated in other areas by previous workers. The lithologies, structures, and grades of metamorphism found on each side of the front are typical for that province and appear quite distinctive. On the "Grenville side" there is a migmatitic terrane is which granite is a major component of the migmatite and which includes generally concordant foliated homogeneous granitic bodies. This terrane is typical of the northwestern part of the Grenville province. From previous studies using the strontium evolution diagram, it has been shown that the determination of a "total rock

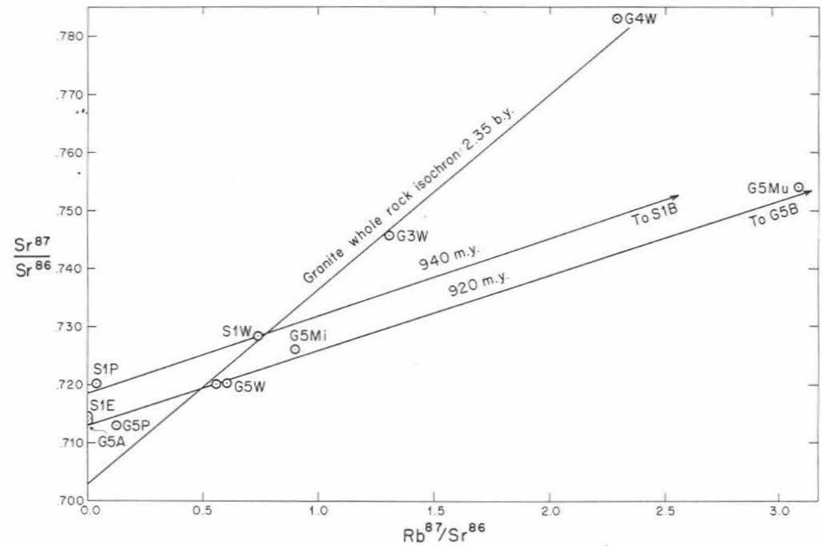

FIG. 5. Mineral and total rock analyses for a granite (G5) and a schist (S1) originally of Superior Province age but "cannibalized" during Grenville metamorphism [Grant, 1964].

isochron" and a "mineral isochron" can be used to determine respectively the primary age of crystallization of a suite of cogenetic igneous bodies and also the time at which they were last recrystallized [Compston, Jeffery, and Riley, 1960; Fairbairn, Hurley, and Pinson, 1961; Lanphere, Wasserburg, Albee, and Tilton, 1964]. Applying this technique to a suite of granitic rocks which were taken from a traverse across the front, Grant found that the total rock samples lie along a fairly well-defined isochron with a slope corresponding to about 2.35 AE (Figure 4). Mineral isochrons for a granite and an adjacent schist well within the Grenville region showed that they had achieved virtually complete Sr isotopic homogenization at about 930 million years ago, indicating this "Grenville" age of metamorphism (Figure 5). A granite north of the front which appeared macroscopically unaffected by the Grenville orogeny shows that a partial strontium isotopic reequilibration took place among the constituent minerals also within Grenville time (Figure 6). This study confirms that granitic rocks and metasediments of the Superior province with primary ages of 2.35 AE or greater were reconstituted during the Grenville orogeny and now form part of the Grenville province. In addition, there is distinct evidence that the Grenville episode produced metamorphic effects within what is macroscopically the Superior province. This study shows that at least part of the younger orogenic belt is older continental material and that the band which defines the Grenville front is, in terms of registering effects of this orogeny, probably several miles wide. A study by Krogh [1964] and Hills and Gast [1963] on Grenville province igneous and metamorphic rocks in the Hastings Basin area and the Adirondacks farther to the southeast has shown that there do not appear in this region any rocks which are older than 1300 million years. In general, it therefore appears that some of the younger material comprising this Grenville orogenic belt is reworked older rocks, and some is "new" material added to the continents. The positive identification which was made of Superior province rocks is more definitive than the results which fail to produce evidence of older ages, and so "new" material must be used with some caution (we will return to this point later). It is clear that more extensive studies across the boundaries of this and similar "orogenic" units will be pursued with great intensity in order to give a more complete answer to these questions. In addition to the obvious important tectonic implications of such studies as the preceding one, it is also clear that these techniques are of fundamental importance

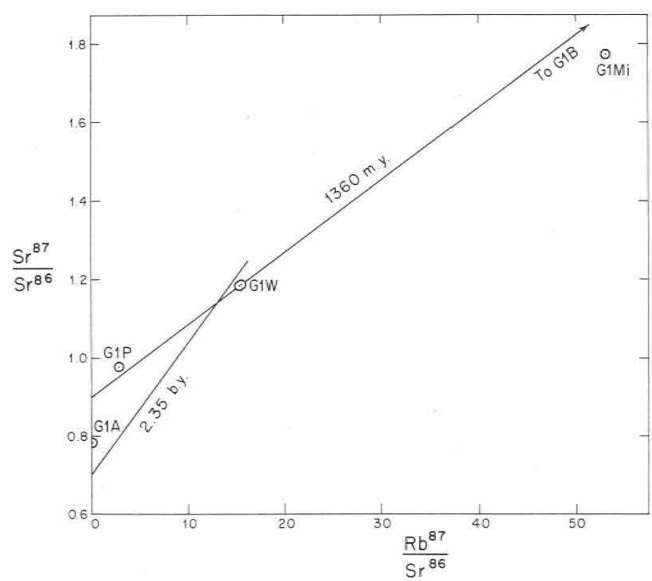

FIG. 6. Mineral and total rock analyses for a granite (G1) in the Superior Province in the neighborhood of the Grenville Front. Note the displacement of the mineral points from the 2.35 AE isochron which shows incomplete Sr isotopic equilibration during Grenville metamorphism [Grant, 1964].

to the field of metamorphic petrology. The use of isotopic redistribution as a means of detecting even very subtle metamorphism and of understanding the nature of the processes which control element redistribution in general will become an inherent part of the subject of metamorphic petrology and *not* an independent activity. Argon loss, lead-uranium diffusion, and strontium homogenization and oxygen isotope ratios will belong in the text of metamorphic petrology along with the other physicochemical methods which have long been a part of the organism of this discipline.

Common Pb and Sr

In the "dating" methods the usual parameters are the amount of daughter isotope produced by radioactive decay and the amount of parent isotope present today. In general the chemical elements which we observe today represent the materials produced during nucleosynthesis which were incorporated in the earth and the contribution from the decay of natural long-lived activities within the earth. The relative abundances of those isotopes which have long-lived progenitors to those which have none is therefore a function of the chemical abundances of parent and daughter, and a function of time. The study of the isotopic abundances of such elements in systems which are not selected because of a high parent/daughter ratio and which have relatively small radiogenic enrichments constitutes the so-called common lead and common strontium problems.

If continental crust, oceanic crust, and the mantle materials have different chemical abundances of daughter and parent elements, the

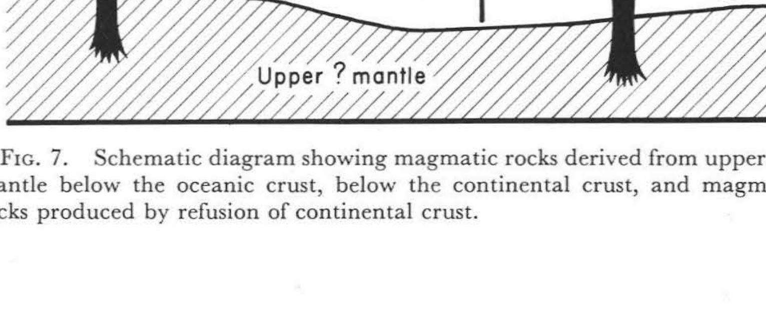

FIG. 7. Schematic diagram showing magmatic rocks derived from upper (?) mantle below the oceanic crust, below the continental crust, and magmatic rocks produced by refusion of continental crust.

study of the relative isotopic composition of common Pb and common Sr gives a means of distinguishing material from these different sources and of following their time evolution and the fractionation of U-Th-Pb, which is so important in the heat balance of the earth (see Figure 7).

After the demonstration by Nier [1938] that the isotopic composition of ore leads was variable and roughly a function of age, and with the

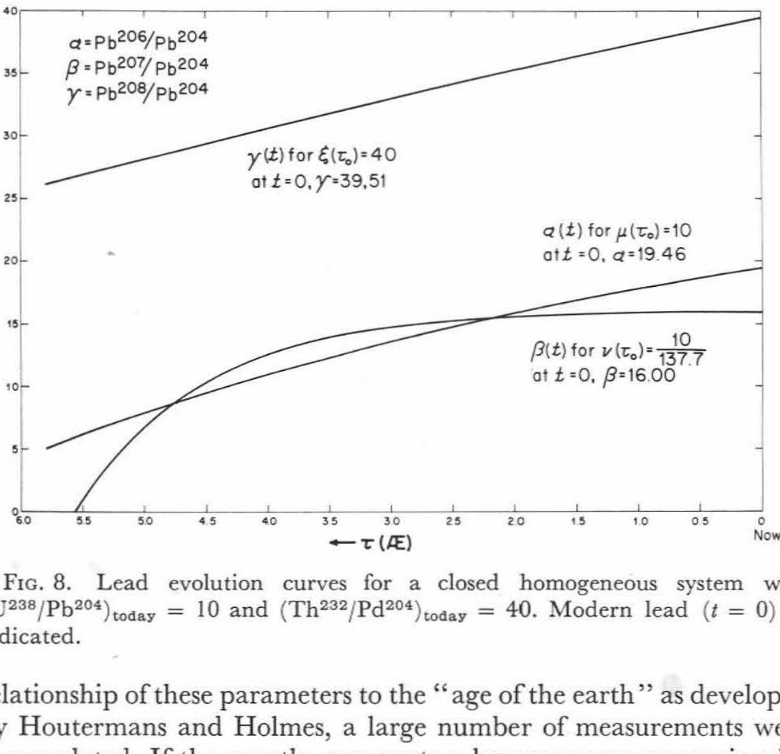

FIG. 8. Lead evolution curves for a closed homogeneous system with $(U^{238}/Pb^{204})_{today} = 10$ and $(Th^{232}/Pd^{204})_{today} = 40$. Modern lead ($t = 0$) as indicated.

relationship of these parameters to the "age of the earth" as developed by Houtermans and Holmes, a large number of measurements were accumulated. If the mantle represents a homogeneous reservoir with given ratios of U^{238}/Pb^{204} and Th^{232}/Pb^{204}, then the relationship between age and isotopic composition will be as shown in Figure 8. This model has been investigated with ore leads by Russell [1963] and Russell and Farquhar [1960], and has been presumed by some workers to represent the evolution of mantle lead. In terms of the lead evolution diagram introduced by Houtermans (Figure 9), the evolution of lead is described by the coordinates $\alpha = Pb^{206}/Pb^{204}$, $\beta = Pb^{207}/Pb^{204}$, $\gamma = Pb^{208}/Pb^{204}$ and are parametrized by the time at which lead is removed from the parent reservoir and the abundance of U^{238} relative to Pb^{204} today (μ).

If the source of magmatic rocks is a homogeneous (U-Pb-Sr-Rb) reservoir in which the ratio of U^{238}/Pb^{204} only changes by the effect of radioactive decay, then samples of these rocks will have as their initial Pb isotopic composition the same value as the homogeneous reservoir at that time (and the same value of μ). They will then lie along a constant μ curve. If there are several magmatic sources which originally had the same Pb isotopic composition but different initial U^{238}/Pb^{204} ratios and which maintained their separate integrity,

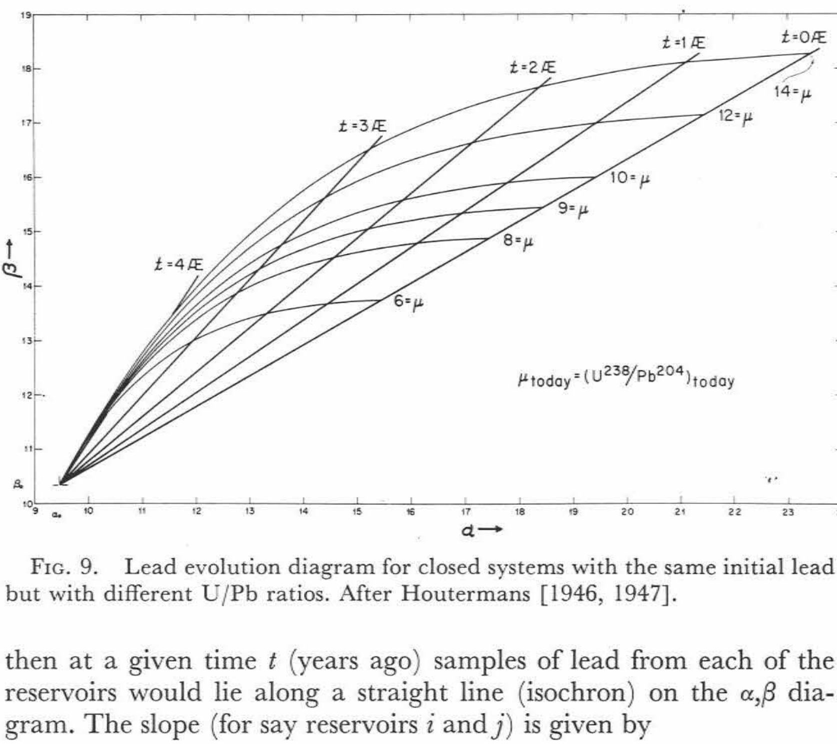

FIG. 9. Lead evolution diagram for closed systems with the same initial lead but with different U/Pb ratios. After Houtermans [1946, 1947].

then at a given time t (years ago) samples of lead from each of the reservoirs would lie along a straight line (isochron) on the α,β diagram. The slope (for say reservoirs i and j) is given by

$$\frac{\alpha_i - \alpha_j}{\beta_i - \beta_j} = \left(\frac{U^{238}}{U^{235}}\right)_{\text{today}} \frac{[e^{\lambda_8 \tau_0} - e^{\lambda_8 t}]}{[e^{\lambda_5 \tau_0} - e^{\lambda_5 t}]}$$

This is a function of the "age" τ_0 of the parent system and the age t. If τ_0 is known, then the time "t" is determined. For modern samples $t = 0$, and it is possible to calculate τ_0. If τ_0 and the initial isotopic composition are known, it is possible to calculate μ from the equation for α given in Table 2. A similar set of equations holds for $\gamma = Pb^{208}/Pb^{204}$ and α which, however, depend on the Th^{232}/U^{238} ratio today.

On this diagram (Figure 9) all unfractionating systems which originally had the same Pb isotopic composition would lie on a straight line

Table 2

EQUATIONS FOR RESERVOIRS CLOSED OR SUBJECT TO LOSS IN WHICH U-TH-PB ARE NOT FRACTIONATED. NOTE THAT $\mu(\tau_0)/\nu(\tau_0)$ IS A CONSTANT (137.8) FOR ALL TERRESTRIAL SAMPLES

$$\alpha(\tau)-\alpha_0=\mu(\tau_0)\left[e^{\lambda_8\tau_0}-e^{\lambda_8 t}\right]$$
$$\beta(\tau)-\beta_0=\nu(\tau_0)\left[e^{\lambda_5\tau_0}-e^{\lambda_5 t}\right]$$
$$\gamma(\tau)-\gamma_0=\xi(\tau_0)\left[e^{\lambda_2\tau_0}-e^{\lambda_2 t}\right]$$
$$\frac{\alpha-\alpha_0}{\beta-\beta_0}=\frac{\mu(\tau_0)}{\nu(\tau_0)}\frac{\left[e^{\lambda_8\tau_0}-e^{\lambda_8 t}\right]}{\left[e^{\lambda_5\tau_0}-e^{\lambda_5 t}\right]}$$

whose slope determines the time since they were isotopically identical. It was exactly ten years ago that C. C. Patterson in his classical experiment showed that the lead from iron meteorites (uranium free), stone meteorites, and "modern" lead from basalts and deep-sea sediments lie on a straight line corresponding to a time of 4.5 AE, a time which is often taken as the "age of the earth." These are the first and most direct data which relate other solar bodies and the earth. Ages of between 4.0 and 4.7 AE were obtained on meteorites by the Ar^{40}-K^{40} and Sr^{87}-Rb^{87} methods. These data established this time as a period of major chemical differentiation in the history of the solar system. The values α, β for Fe meteorites are taken to be the isotopic composition of lead at the time of formation of the earth.

In studying ore leads (and a suite of volcanic leads), it was found that some were anomalous in that they lie in the region to the right of the "modern isochron" and would yield negative (i.e., future ages) in terms of the parameters used for α_0, β_0, and τ_0. This leads to the question of the correctness of α_0, β_0, and τ_0 and whether or not it is possible to assign characteristic values of μ to lead from various sources. This problem has been under attack for the past few years by studying primary Pb in U-free minerals in magmatic rocks whose age of crystallization is known (from independent isotopic age measurements on other phases or using other decay schemes) and by studying the lead isotopes in oceanic sediments and beach sands [Patterson, 1964]. Earlier studies in which it was not possible to assign an age independent of the common Pb composition made it difficult, if not impossible, to determine whether Pb was "anomalous" in terms of the simple model of growth in a nonfractionating reservoir.

Houtermans' model is very simple in that it assumes that U and lead in particular are not fractionated during the formation of crustal materials. This is not demonstrable independently and is best determined by testing the model on well-understood systems. An example of such an investigation is the recent work of Zartman on lead in plutonic and metasedimentary rocks of the Llano region in Texas, part of the "Grenville" age province. The igneous rocks have been well dated by the major methods. Primary lead in the feldspars (which are essentially U free) from all the principal igneous rock types showed a rather uniform isotopic composition and gave model ages t (using the constants α_0, β_0, τ_0 given by Patterson) which are in good agreement with the ages determined by other methods and on more radiogenic minerals. The relative uniformity of the primary lead suggests a homogeneous source. The measured values of μ for the total rocks and the calculated values of μ from the model are in good agreement, indicating no serious Pb-U fractionation in the formation of the granites and the parent material in this particular case.

Such work, if extended to other igneous bodies of the same age, will determine whether all of these approximately contemporaneous igneous rocks came from the same magma source or from different sources. On the other hand, the Pb from the metasedimentary rocks was isotopically very variable and confirmed the complicated origin which is apparent from geological considerations. A study by Catanzaro and Gast [1960] indicated rather good agreement between model ages and the regular ages for some pegmatites as old as 2.7 AE. From the few published data, it appears that the simple model is grossly correct and that α_0, β_0, and τ_0 cannot be greatly different from the values given by Patterson.* However, such studies are still not complete, and it is particularly important that they be extended to more well-dated plutonic rocks.

The consequences of fractionation between Pb and U in the formation of magmatic bodies can be considered in terms of several exchanging homogeneous reservoirs. This has been approached by Patterson from consideration of the lead in beach sands and deep-sea sediments. A theoretical discussion of the simple two-reservoir case is of interest. The equations are given by Wasserburg [1964]. The solution for a simple two-layer model with constant transport coefficients are shown in Table 3.

* The work of Reed, Kigoshi, and Turkevich [1960] on the U, Th, and Pb concentrations in stony meteorites shows that there remains some difficulty in explaining the material balance in the parent-daughter systems for these bodies. This observation should not be obscured by our enthusiasm for "magic numbers."

Table 3

SOLUTIONS FOR α AND β FOR CRUST (1) AND MANTLE (2) FOR THE CASE OF A CONTINUOUSLY GROWING CRUST.

Crust	Mantle
$\mu_p^1/\mu_p^\Sigma = \dfrac{(1-e^{-H\tau})}{(1-e^{-G\tau})}$	$\mu_p^2/\mu_p^\Sigma = e^{-(H-G)\tau}$
$\alpha^1 = \alpha_0 + \mu_0 \dfrac{\left[1-e^{-\lambda_8\tau} - \dfrac{\lambda_8}{(G-H-\lambda_8)}(e^{-(\lambda_8+H)\tau} - e^{-G\tau})\right]}{(1-e^{-G\tau})}$	$\alpha^2 = \alpha_0 + \dfrac{\mu_0\left[e^{-(\lambda_8+H-G)\tau} - 1\right]}{(G-H-\lambda_8)}$
$\beta^1 = \beta_0 + \nu_0 \dfrac{\left[1-e^{-\lambda_5\tau} - \dfrac{\lambda_5}{(G-H-\lambda_5)}(e^{-(\lambda_5+H)\tau} - e^{-G\tau})\right]}{(1-e^{-G\tau})}$	$\beta^2 = \beta_0 + \dfrac{\mu_0\left[e^{-(\lambda_5+H-G)\tau} - 1\right]}{(G-H-\lambda_5)}$

G and *H* are the transport constants for lead and uranium, respectively. The symbol Σ indicates the value for the total system $(1 + 2)$, and the subscripts *p* and *o* indicate present and original values, respectively.

Here the functions G^j and H^j are the fractional rates of transport of Pb^{204} and U^{238} out of reservoir *j* and represent the time constants for chemical differentiation of the earth. If the transport is solely from mantle to form crust, then

$$H^{\text{mantle}} = \frac{-\mathscr{E}}{V^{\text{mantle}}} \frac{dV^{\text{mantle}}}{dt}$$

where $\mathscr{E}$ is the enrichment factor for uranium of the magma compared to the mantle, and $(1/V^{\text{mantle}})(dV^{\text{mantle}}/dt)$ is the fractional rate of change of the volume of the mantle. A schematic drawing of this is illustrated in Figure 10. It is possible to estimate this parameter from considerations of crust and mantle volumes and the estimated content of U in these regions. It then follows that α and β for the total crust depend not only on the time but on G^{mantle} and H^{mantle}. For the mantle and hence for the materials added to the crust from the mantle α and β only depend on G^{mantle}-H^{mantle}, and so a mantle reservoir with different time constants *G*, *H*, but the same value *G-H* will yield leads of identical composition. An evolution curve for the mantle (and crust) is similar in form to the nonfractionating evolution model. In fact a mantle consisting of a family of systems with the same value of *G-H* but with different μ values will also generate isochrons, the only difference being that the slopes will depend on *G-H*, and hence the ages and μ values calculated assuming the nonfractionating model will be in error (Figure 11*a*, *b*). Independent age assignments are therefore absolutely necessary to understand the Pb isotopic evolution. The extent to which this type of fractionation is important can only be answered by more precise data of the type mentioned earlier. The value of τ_0, the so-called age of the earth, is also tied up in these parameters so that the effects of fractionation are important for a number of reasons. The variety of important work currently

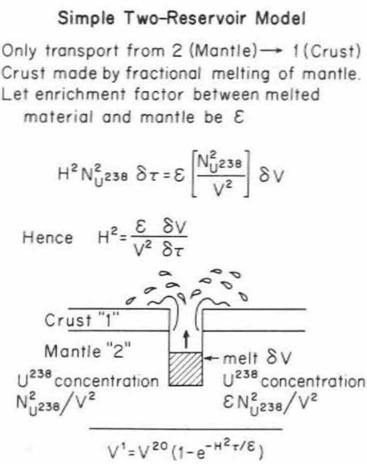

FIG. 10. Relationship between transport constant, chemical fractionation factor, and rate of growth of crust.

taking place in this field is represented by the symposium on lead isotopes at the National American Geophysical Union meeting of 1964.

The evolution of strontium may also be applied to the study of crustal evolution [Gast, 1960; Hurley *et al.*, 1962]. Rb-Sr age determinations on meteorites [Schumacher, 1956; Herzog and Pinson, 1956; Webster, Morgan, and Smales, 1957; Gast, 1962] have shown that there is considerable enrichment in Sr^{87} in chondrites due to Rb^{87} decay. The enrichment observed in primary strontium in terrestrial igneous rocks is much lower as shown in Figure 12. This means that the ratio of Rb/Sr for the source of these materials is considerably lower than for chondritic meteorites as emphasized by Gast [1960]. In terms of the evolution of strontium in the mantle as determined on rocks of supposed mantle origin, this means that the total enrichment is at most only 1.4 per cent (and most probably between 0.7 and 1 per cent) over a period of about 4.5 AE or about 0.3 per cent per AE. The present precision of good quality solid source mass

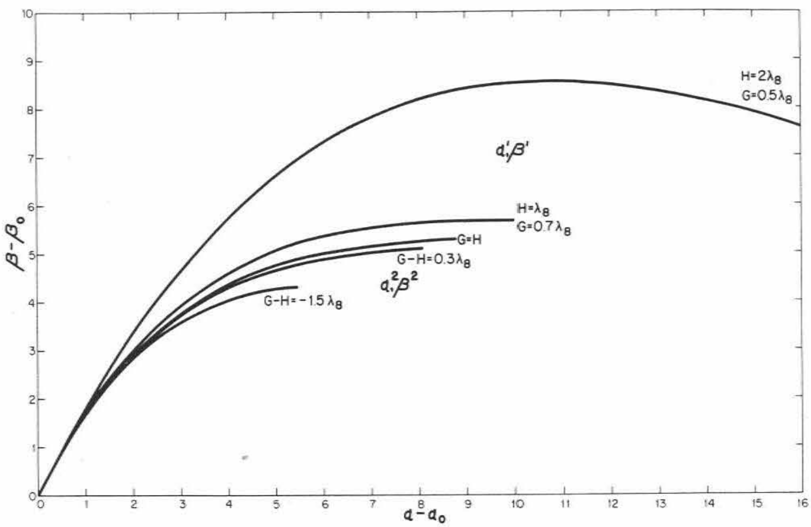

FIG. 11(*a*). Lead evolution curves for crust (1) and mantle (2) for two-layer model with transport constants G and H. It is assumed (without justification) that the crust is enriched in uranium with respect to lead. The composition of lead added to the crust at a given time is read off of the mantle curve for the appropriate time.

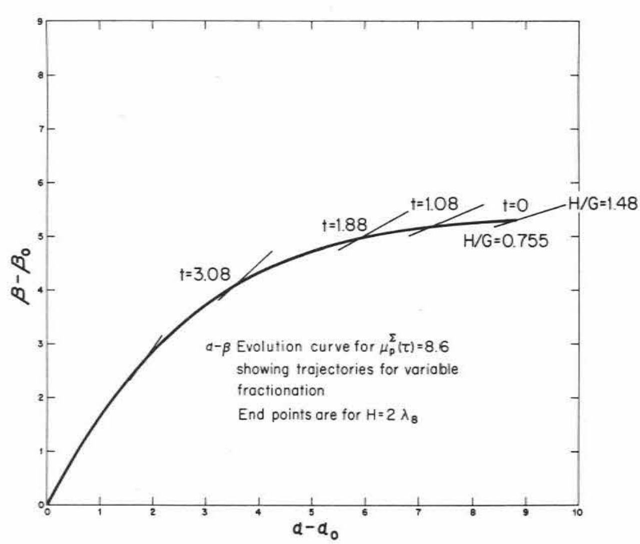

FIG. 11(*b*). Lead evolution diagram showing growth curve for total system and trajectories for variable U-Pb fractionation (H/G) at different times.

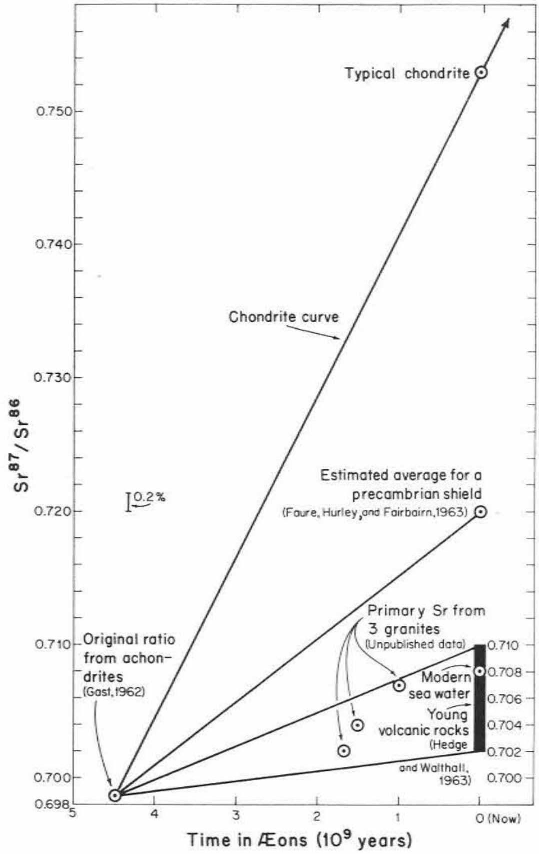

FIG. 12. Schematic diagram for Sr^{87}/Sr^{86} growth with time for chondrites showing the comparison with modern sea water, young volcanic rocks (continental and oceanic) and a present-day shield area.

spectrometric measurements is about 0.1 to 0.2 per cent and hence the detailed exploitation of Sr isotopic evolution is dependent on a refinement in technique. The enrichment in crustal Rb/Sr over that found in presumed mantle material can permit the distinction between remelted crustal material and the addition of new mantle material. The distinction between primary mantle material and refused crustal material can be very positive in the case that the refused crustal material has a long enough history in a Rb rich environment. However, in the case that this ratio is not high enough, the distinction cannot be as strong since the great heterogeneity of crustal materials can certainly provide sources with a variety of Rb/Sr ratios.

With regard to the question of normal strontium, primary Sr^{87}/Sr^{86} values for some granites are typically from 0.702 to 0.708, whereas young basic volcanic rocks have typical ratios of from 0.702 to 0.707. Estimates of the mean Sr^{87}/Sr^{86} ratio in the Precambrian shield of N. America (Gast, Faure, Hurley, and Fairbairn) indicate this to be ≈ 0.72. The refusion of such shield materials today would produce magmas with an original strontium quite different from the above-mentioned range of values and would indicate a crustal origin. This has already been used on metasedimentary rocks and is clearly an indication of crustal history. The clarification of the time evolution of Sr and the Rb/Sr ratio is being pursued by investigations on Sr rich-Rb poor minerals from rocks with simple histories and of known age.

As mentioned earlier, the assumption of a simple homogeneous mantle reservoir for continental volcanic rocks is uncertain because of the possibility of crustal contamination, and it would appear that the oceanic volcanic materials would yield better samples of upper crustal material. A very recent study by Lessing and Catanzaro [1964] on Hawaiian trachytes, and a study by Gast, Tilton, and Hedge [1964] on the isotopic composition of lead and strontium in several volcanic rocks from Ascension and Gough Islands on the mid-Atlantic ridge showed that this simple assumption is invalid. They reported that Sr^{87}/Sr^{86} varied for different samples from the same island and between two islands. The magnitude of this effect is about 0.7 per cent and would constitute half of the total effect due to mantle evolution as estimated earlier. They also reported large isotopic variations of α and β for samples from the same island and between the islands. The samples from Gough Island fall very close to the modern model isochron but have differences of isotopic composition

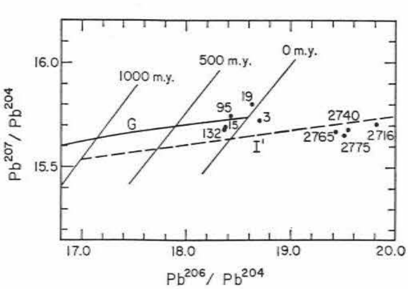

FIG. 13. Part of a lead evolution diagram [parameters after Murthy and Patterson, 1962], showing the experimental points for Gough and Ascension Islands [Gast, Tilton and Hedge, 1964].

which would correspond to model age differences of almost 250 million years. The latter effect is present in the Ascension Island samples, but they would yield negative model ages of up to 700 million years as shown in Figure 13. Anomalous leads and strontium abundances have been observed previously in continental areas, but this result emphasizes that the assumption of a homogeneous upper mantle in oceanic areas is not justified and that some of the anomalies observed in continental areas may not so obviously be used as an indication of crustal contamination. Petrologic investigations have indicated that it is necessary to assume two magma sources in order to explain the extrusives in Hawaii. The isotopic data confirm this minimal degree of complication and demand isotopic as well as chemical heterogeneity on what appears to be a local scale for significant time periods. Such different sources are apparently mechanically available for extrusion over extremely short time periods.

The results on oceanic leads and Sr indicate Pb-U-Th and Sr-Rb fractionation during crustal evolution and suggest more complexities than are even hinted at by a simple fractionating two layer crustal-mantle model. Clarification of such phenomena will demand the interest and activity of all the branches of the earth sciences.

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